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Bunched Release of Gases from Oxide Targets

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Abstract

Targets made out of oxides of the alkaline earth metals have been shown at ISOLDE to be among the fastest targets for production of radioactive beams of the rare gas elements. In addition oxide target materials release nitrogen and seem to be the only ones which may release the element carbon due to its oxidation to the high temperature stable CO gas. Thick targets of MgO and CaO have been studied in connection with their use at the 1GeV pulsed proton beam from the PS-BOOSTER synchrotron. It is shown that the 2.4 μs short proton pulse causes a pronounced bunched release with short delay of these elements which gives rise to 100-200 ms wide pulses of radioactive ion beams. Pulse shapes and overall yields of ion beams of He, Ne and Ar isotopes are studied as function of temperature and proton pulse intensity. For carbon and nitrogen also the release as function of the observed ionic species C^+, CO^+, N^+ and N_2^+ is discussed.

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1. Introduction

1.1 Background

During more than 30 years, there has been a continuous search for target materials with fast release properties for the production of short-lived "exotic" nuclides at the rims of the proton vs. neutron nuclear chart. In the later years, groups of target materials serving the production of specific nuclear reaction products have been developed at ISOLDE [1-4].

1.2 ISOLDE at the PS BOOSTER

At the new location, ISOLDE is getting 1 GeV protons, slightly higher than the 600 MeV at the synchrocyclotron (SC). However, an extremely important difference between ISOLDE at the SC and the new installation at the PS- BOOSTER synchrotron is that the proton beam is strongly bunched [5]. While the average proton current in the new location is the same as before or slightly lower the instantaneous intensity of the proton beam is increased by a factor of 1000.

The resulting instantaneous heating has also proven to be very advantageous not only for studying the performance of targets but also for the production of very short-lived nuclides far from stability since it together with the increased formation cross-sections enhances their yields.

Several different oxides were tested and run at the SC [1-3]. At the present ISOLDE installation only one magnesium oxide, MgO, target and four calcium oxide, CaO, targets have been used and their performances have been tested in connection with the production runs for physics.

2. Target materials

The ISOLDE target container and oven, see e.g. [6], is made of tantalum metal in the form of a tube with a product exit tube connecting it to the ion source. In order not to react with the tantalum all the oxide materials are filled into rhenium boats placed inside the tantalum tube. The MgO target was made from commercially available oxide. This was first heated in a porcelain crucible at 800^0C for a few hours and then transferred to a Re-boat for further heating at 1400^0C. After some hours the MgO powder has sintered and its volume decreased to about 1/3 of its original volume. More of the dried oxide is then filled on top of the already sintered MgO and reheated. This procedure is then repeated until the desired volume in the boat is reached.

This procedure does not give the most open structure of the target material, but gives a target material that is very stable. Heating to 1500^0C for 3 hours gave no significant weight loss. The thickness of this target was 2.9 g/cm^2.

The targets of CaO were made from commercially available $CaCO_3$ which was first thoroughly dried by heating in a porcelain crucible for 4 hours at 600^0C and followed by 14 hours at 200^0C. A Re boat was then filled with this dried calcium carbonate which was decomposed in vacuum to 1000^0C for

about 48 hours. Contrary to the MgO material, the CaO powder was white, fluffy and only little sintered. The target thickness of the CaO targets were fairly equal and about 4.1 g/cm^2.

3. Target performance

The target performance used to be synonymous with the data printed in the ISOLDE User Guide [7] which gives the observed atoms per second for a primary proton beam of 1 μA. These data were measured for all the nuclides produced at the former installation. Measurements of the production rates at the SC were very simple to perform since the proton beam as well as the radioactive beams were close to DC currents.

At the BOOSTER the oxide targets did not suffer too much the deleterious effects of the beam power density but operated successfully up to 13 days and kept their best performance for at least 48 hours.

A decidedly positive effect of the separated bunches of protons is the possibility to measure nuclides with short half-lives (<1s) in very good signal to noise conditions. Also, it seems to be experimentally confirmed that the high instantaneous proton pulse has the positive effect to release the products from the target material in a bunched form. There may be different explanations to this effect like shock waves or simply enhanced diffusion due to local extra strong heating cycles. Measurements of the radioactive ion-beam intensity as function of time i.e. the pulse shape or release function and yields of the oxide targets will be the subjects of the present paper and the questions of mechanisms giving these results will be discussed. In order to determine the total number of nuclides arriving at the measuring station per proton bunch the complete release-rate curve has to be known and integrated. The parameter definitions and the numerical treatment of these curves and their integration is treated by Lettry et. al. [8].

Results obtained for release times and yields of He and Ne by use of the MgO-target were few and very similar to results from CaO. Only results from runs with CaO are treated in the following.

3.1 Product release

Even the very first published information on the ISOLDE project [9] did contain information on average delay times for different targets. Present day ISOLDE technology makes it both possible and needed not only to give average delay times, but to give the complete time distribution of the intensity of arriving products to the measuring stations. The oxide targets are particularly successful in the respect that the shortest delays from the time of the proton impact on the target to the maximum of the release curve are obtained with these target materials.

3.1.1 Noble gases

Typical release curves are shown in Fig.1. By fitting an empirical curve to the data the total yield of ions per proton pulse can be obtained by integration as discussed in [8].

Note that the curves discussed in this paper are $P_i(t)$ as defined on ref. [8] the measured count rates and therefore depends on the half-lives of the nuclides so that the release of a nuclide with a short half-life would seem to be faster than a nuclide with a long half-life. By increasing the temperature we would expect that the release becomes faster. Another parameter of importance for the transport time is the mass of the nuclide or molecule transported. In Fig. 1 we see that at least for the noble gases this is much more important than the temperature. Thus, Fig. 1 shows both the result of increasing the target temperature from 950 to 1340^0C for argon and the release curves for Ne and He at 950^0C.
The fall time deduced from the fitted P(t) curves can be shown to be very strongly correlated with the calculated time, t_{50} for 50 per cent of the activity to reach the measuring station. We thus show t_{50} as a function of the temperature for all the measurements done on Ar-isotopes in Fig. 2. In this figure the values are all corrected to give the time for stable nuclides. Contrary to our early expectations, the measurements show that t_{50} is largest at the highest temperature.

3.1.2 Nitrogen

One of the very difficult elements to produce efficiently on-line is nitrogen. Targets of graphite [10], Platinum-graphite [11] and Sodium-zeolite [12] have been used. As oxygen is a very suitable target element for the production of many nitrogen isotopes by (p,2pxn) and (p,αxn) reactions, a good possibility is using the oxide targets.
Both monatomic N^+ and diatomic N_2^+ beams in the form of $^{14}N^{17}N^+$ were studied and the release curves are shown in Fig. 3. The number of counts for N^+ are multiplied by five in order to emphasise the differences in the curve shapes for the two species. There seems to be a pronounced chemical effect which makes the once formed N_2 molecules more inert against reactions with target materials and with the walls of the tantalum target container and transport line, thus getting a fast noble gas like passage to the ion source. Attempts to optimise the formation of N_2 by addition of $^{14}N_2$ to the target proved to be particularly beneficial as discussed in [10].

3.1.3 Carbon

This element has been even more difficult than nitrogen to separate on line in pure form. Carbon makes very stable compounds with a large number of elements and trace amounts like nuclear reaction products are easily adsorbed by even the smallest amounts of impurities in the target materials. Stable carbides are also formed with tantalum, the most common of target construction materials. The possibility that carbon isotopes produced in an oxide target matrix would form gaseous compounds with the oxygen in this matrix was studied, and the release curves observed for monatomic C^+ and diatomic CO^+ are shown in Fig. 4.
Similarly to the discussion under nitrogen we find that the molecular ion CO^+ is much faster released than the monatomic C^+ ion. In this case the C^+ beam was so weak that one second collection time had to be used in order to get reasonable counting rates. The deduced t_{50} values were also very different with 63 ms for CO^+ and 309 ms for C^+. The t_{50} for CO^+ is unexpectedly short, but that CO as a gas should be released much faster than monatomic carbon was expected.

3.2 Product yields

Yields were given traditionally as radioactive beam intensity in atoms per second produced by irradiating the target with 1μA (particle) protons or other ions. In the bunched beam this is equivalent to calculating the integral of the release curve obtained from a proton pulse of 1μC of protons. By integration of the release curves yields results shown in Fig. 5 are obtained. Included are also results from the SC-installation.

The best studied products are by far the Ar-isotopes and it is possible to compare the results to estimates made from the formulas by Silberberg and Tsao [13]. In Fig. 6 we have given experimental results for two different temperatures and compared them with the results from calculations. Measurements and calculations give in this case comparable results. However, the high temperature yields are much higher than the yields at lower temperature and those results show also most similarity with the calculations.

4. Other oxide targets

At the SC, ThO_2 was used for production of Rn and halogens. ZrO_2 was particularly successful as it was possible to produce Se beams as the molecular ion $SeCO^+$ and BaO for production of Xe. For the production of light Kr isotopes SrO should be an attractive material. Off-line studies of Ta_2O_3 [14] indicate that it will be very suitable for the production of the heavy C isotopes since the CO partial pressure above a mixture of Ta_2O_3 and C is particularly high.

5. Discussion

5.1 Target materials

The main results of the experiments are the following:

1) We did not observe faster release of noble gases when the temperature was increased above 950^0C. On the contrary, the release became slower. This result was unexpected but may be explained by increased sintering of the material at the higher temperature. An alternative explanation is that at low temperature, the active target is only the centre of the heated beam path, but when more external heating is applied, products first diffusing away from the beam path may also arrive at the ion source.

2) The yield of the Ar-isotopes increased when the temperature was increased up to 950^0C. This result seems to support the alternative explanation for the slower release given above, as we do not lose the products moving towards the colder regions. They are only more delayed.

3) No significant effect could be seen by varying proton beam intensity.

5.2 Products

5.2.1 Noble gases

Helium, neon and argon were studied and the release curves for these products in Fig. 1 show that He is released fastest and Ar slowest of these. However, all three show fast release and at temperatures of about 950^0C, 50% of the integrated yield is collected in 134, 441 and 770 ms respectively. Calculated for stable nuclei this will be 177, 450 and 1012 ms.

The comparison of the yields with estimates using the formulas of Silberberg and Tsao show that the efficiencies for Ar-production is of the order of 10%, but for both Ne and He the efficiencies are only of the order of 0.5%.

There is not sufficient data to point at other significant differences between the studied noble gases. A tendency for Ne to show an increased release rate for increasing temperature should, however, be noted.

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Figure captions

Fig. 1. Measured release curves for ^{35}Ar at 950^{0}C and 1320^{0}C, for ^{19}Ne at 950^{0}C and for ^{6}He at 950^{0}C. Target CaO.t_d is the time from proton impact to start collection of the nuclide and t_c is the collection time.

Fig. 2. The half-life corrected release time (t_{50}) for the first 50% of the total collected number of Ar nuclides from all experiments with CaO targets at the present ISOLDE facility.

Fig. 3. Experimental release curves for the two nitrogen species $^{17}N^{14}N_2^+$ and $^{17}N^+$. In order to emphasize the shape differences, the counts for N^+ have been multiplied by five. t_d and t_c: see caption to Fig. 1.

Fig. 4. Experimental release curves for the ions $^{10}C^+$ and $^{10}C^{16}O^+$. A collection time (t_c) of one second was used for C^+. t_d: see caption to Fig. 1.

Fig. 5. A collection of yields from CaO targets at the present ISOLDE. Yields have been measured for ^{6}He, ^{10}C, ^{17}N, ^{18,19}Ne, $^{33\text{-}35}$Ar and $^{41\text{-}46}$Ar. For comparison, values from one target used at the previous installation are included.

Fig. 6. Yields measured at two different temperatures from the same target. Comparison is made with calculations based on the formulas of Silberberg and Tsao [13].

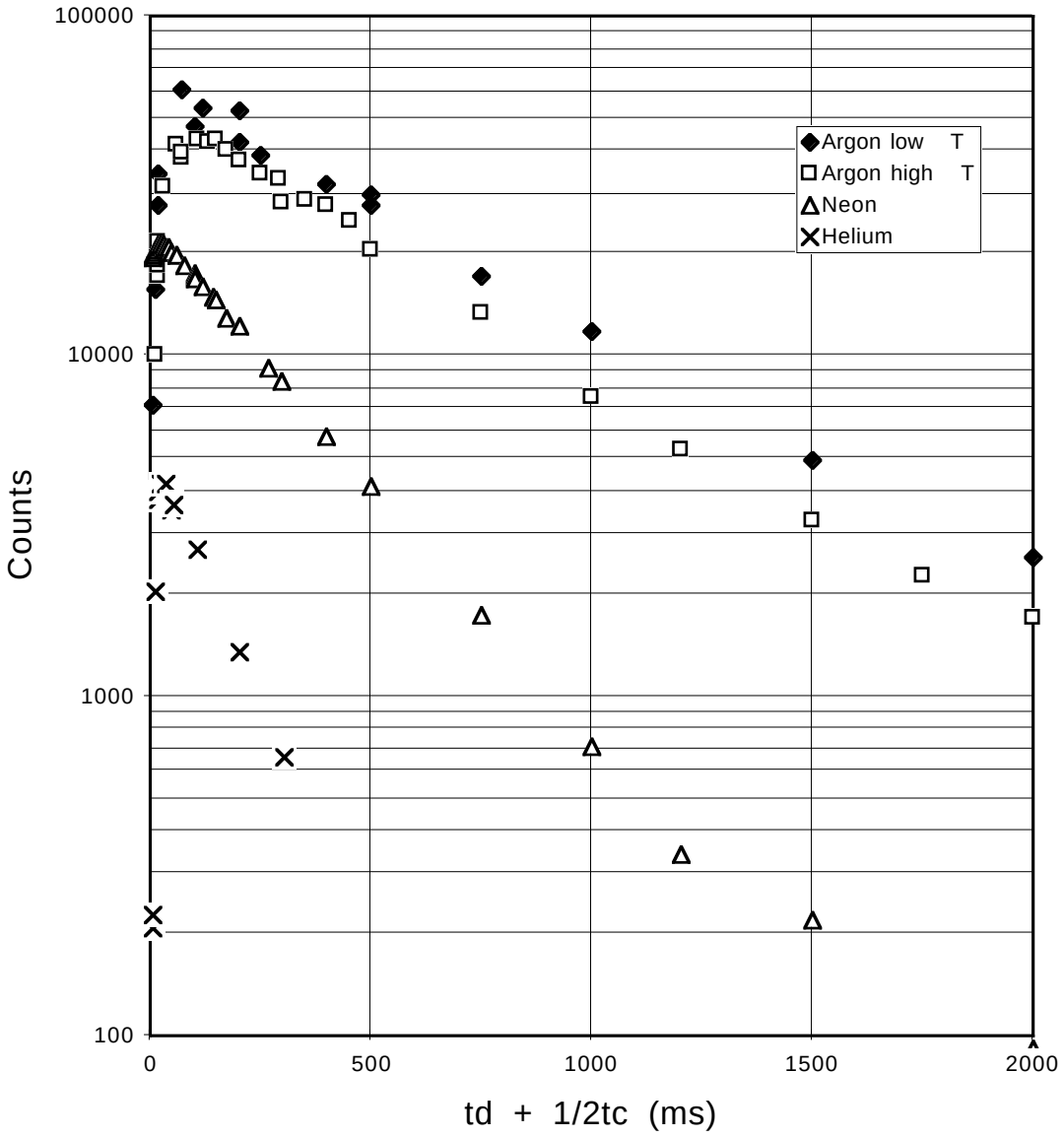

100000
10000
1000
100
Counts
0
500
1000
1500
2000
td + 1/2tc (ms)
Argon low T
Argon high T
Neon
Helium

Fig. 1

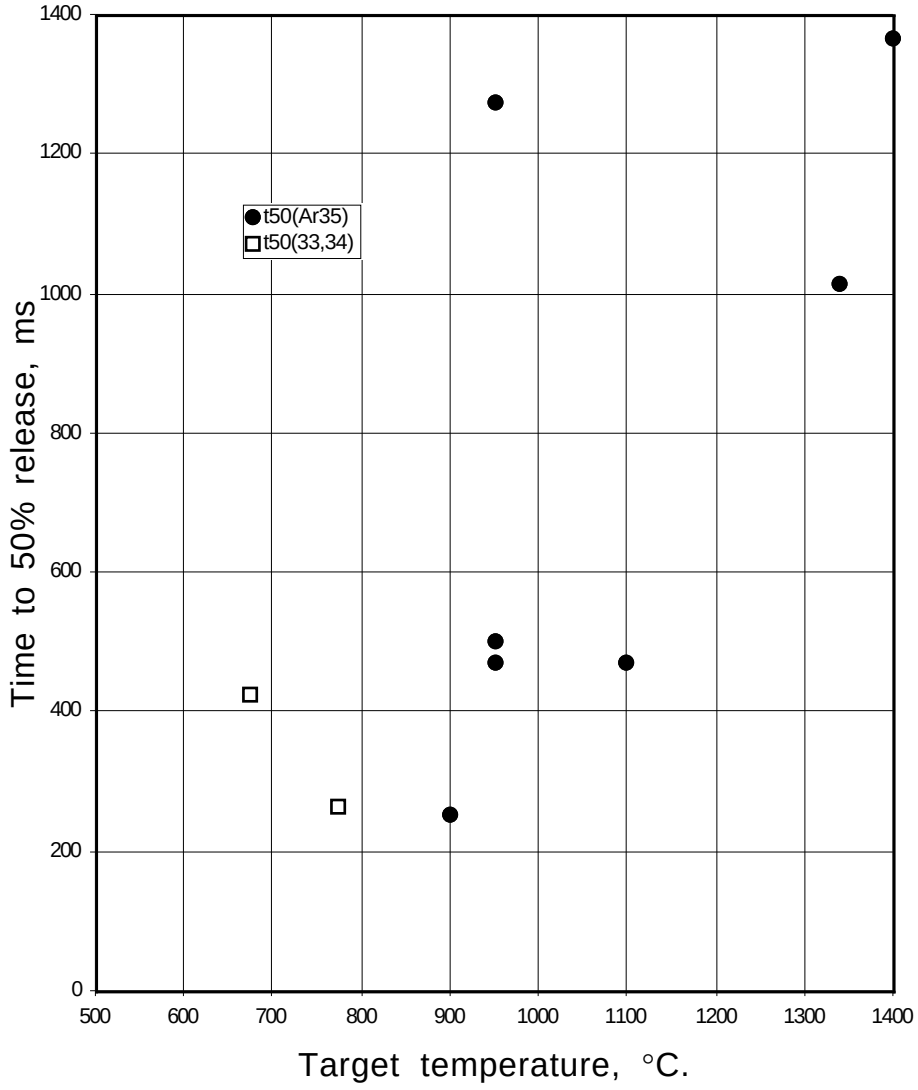
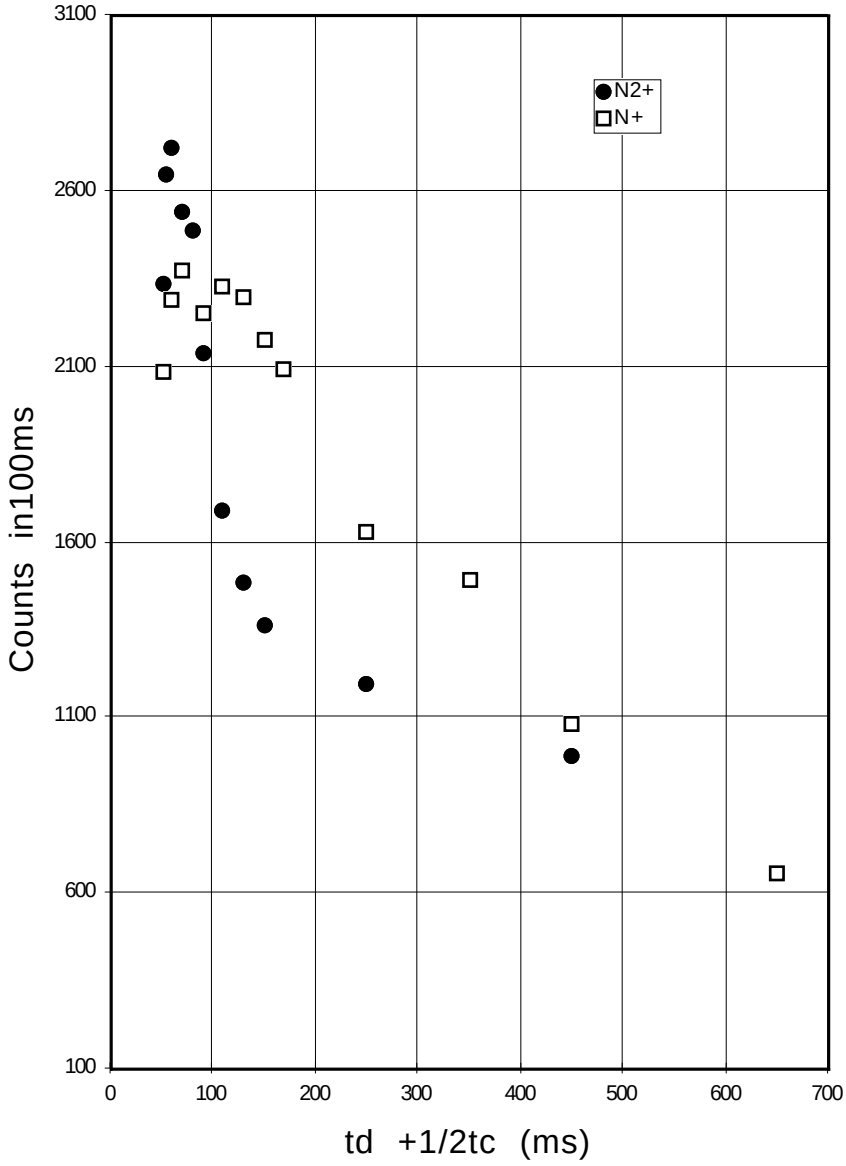

1400
1200
1000
800
600
400
200
0
t50(Ar35)
t50(33,34)
Time to 50% release, ms
500
600
700
800
900
1000
1100
1200
1300
1400
Target temperature, °C.

Fig. 2


3100
2600
2100
1600
1100
600
100
Counts in100ms
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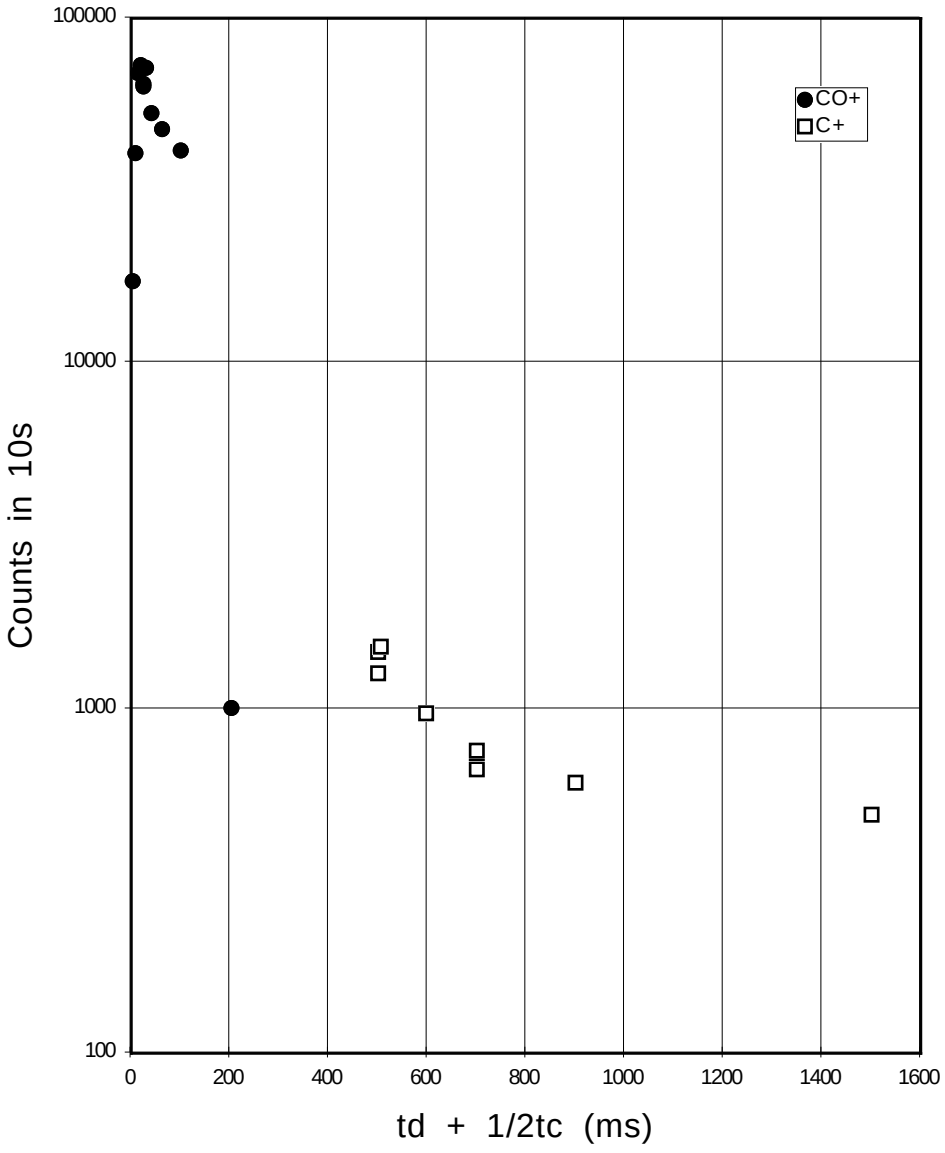
100
200
300
400
500
600
700
td +1/2tc (ms)
N2+
N+

Fig. 3


100000
10000
1000
100
Counts in 10s
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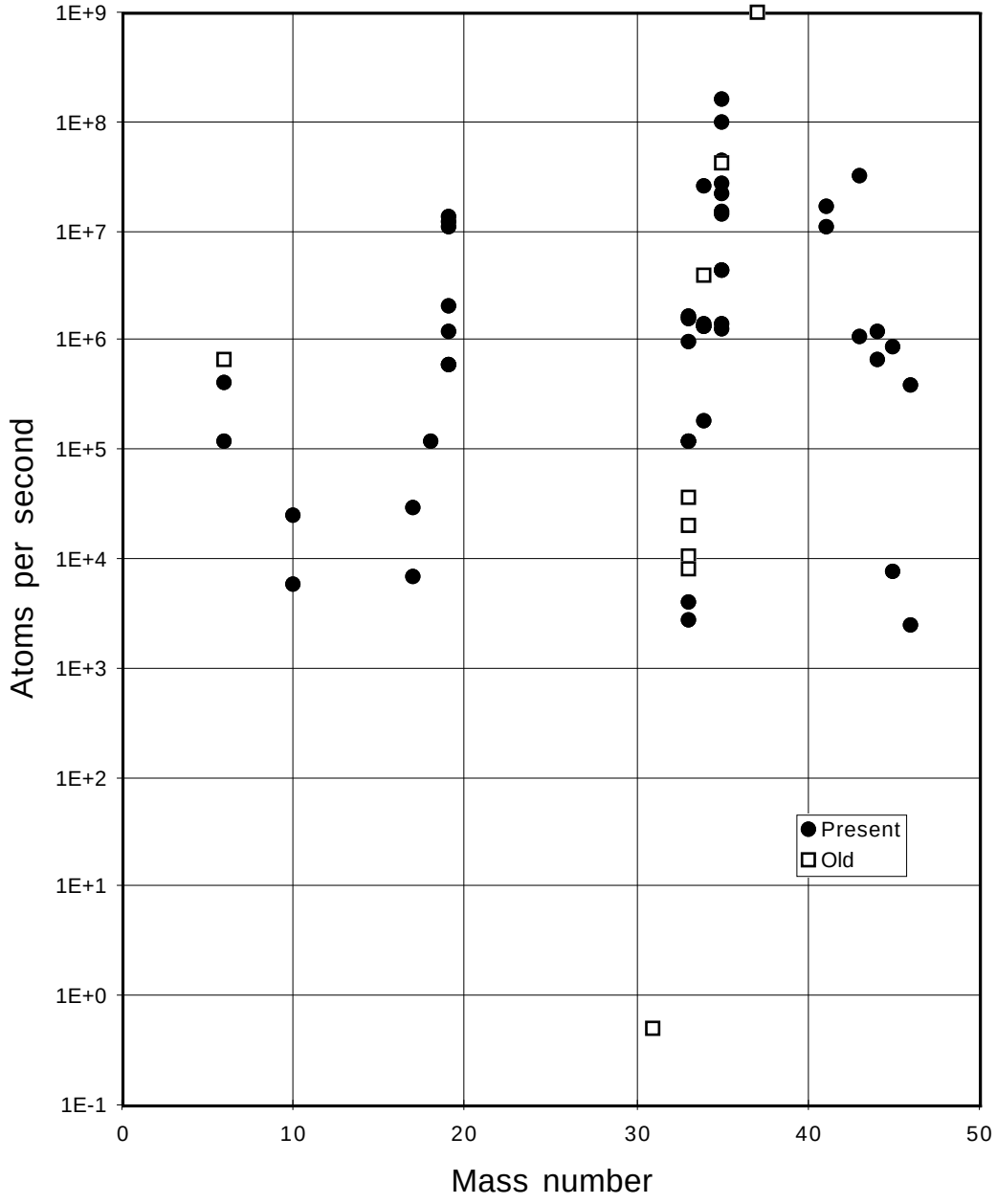
200
400
600
800
1000
1200
1400
1600
td + 1/2tc (ms)
CO+
C+

Fig. 4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1E+8
1E+7
1E+6
1E+5
1E+4
1E+3
1E+2
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1E-1
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10
20
30
40
50
Mass number
Present
Old

Fig. 5

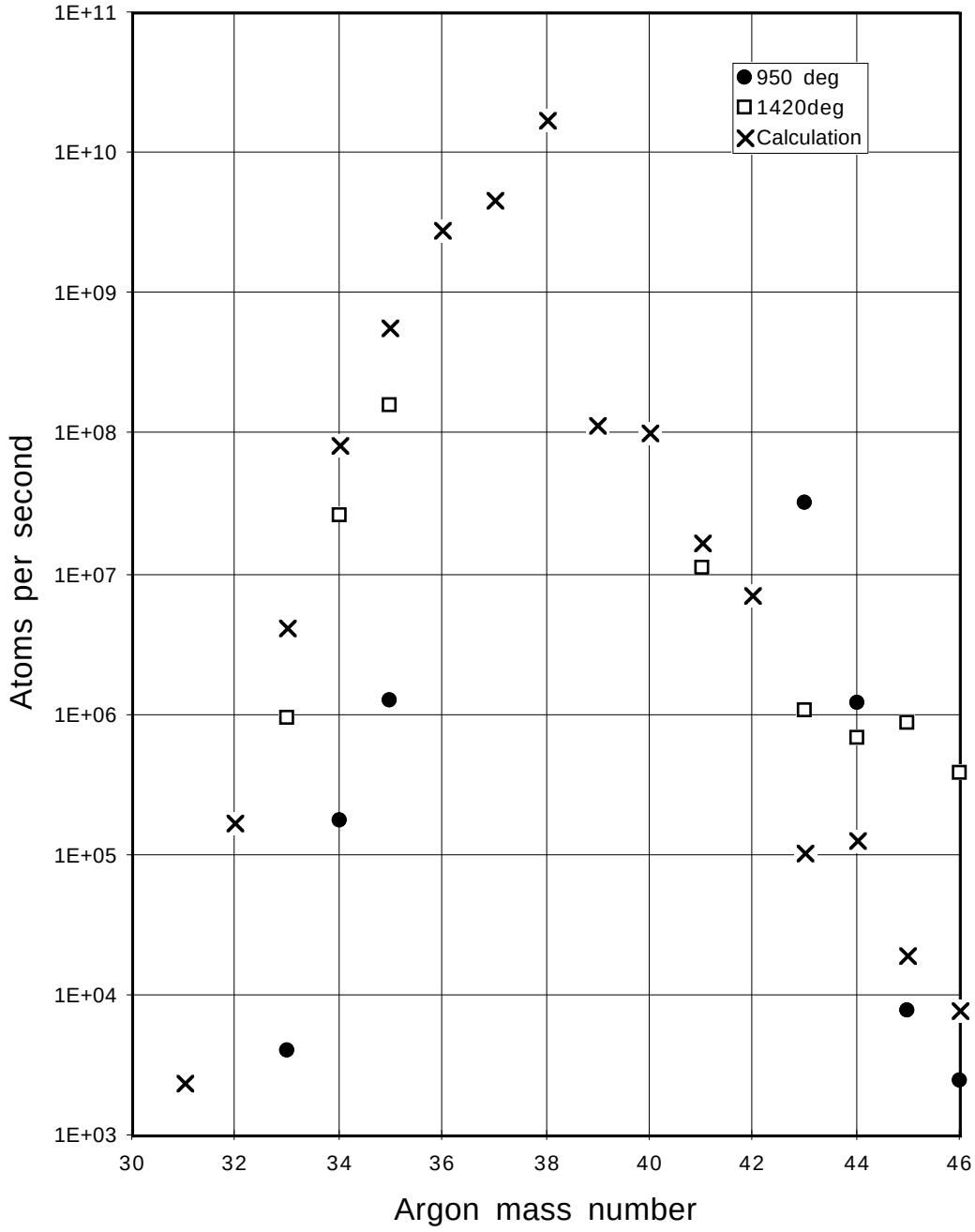
1E+11
1E+10
1E+09
1E+08
1E+07
1E+06
1E+05
1E+04
1E+03
Atoms per second
30
32
34
36
38
40
42
44
46
Argon mass number
950 deg
1420deg
Calculation

Fig. 6